

DOCKET NO: 239016US0



IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :
OTTO MACHHAMMER, ET AL. : EXAMINER: OH, T. V.
SERIAL NO: 10/815,873 :
FILED: APRIL 2, 2004 : GROUP ART UNIT: 1625
FOR: HETEROGENEOUSLY :
CATALYZED PARTIAL DIRECT
OXIDATION OF PROPANE AND/OR
ISOBUTANE

APPEAL BRIEF

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

This is an appeal of the Final Rejection dated February 27, 2007. A Notice of Appeal is submitted herewith.

I. REAL PARTY IN INTEREST

The real party in interest in this appeal is BASF Aktiengesellschaft, having an address at Ludwigshafen, Germany 67056.

II. RELATED APPEALS AND INTERFERENCES

Appellants, Appellants' legal representative and the assignee are aware of no appeals, interferences, or judicial proceedings which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

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III. STATUS OF THE CLAIMS

Claims 1-32, all the claims in the application, stand rejected and are herein appealed.

IV. STATUS OF THE AMENDMENTS

No amendment under 37 CFR 1.116 has been filed.

V. SUMMARY OF THE CLAIMED SUBJECT MATTER

Independent Claim 1 is a process for heterogeneously catalyzed partial direct oxidation of propane and/or isobutane to at least one of the target products acrylic acid, methacrylic acid, by feeding a starting reaction gas mixture comprising propane and/or isobutane, molecular oxygen and at least one inert diluent gas and having an inlet pressure P^1 to a reaction stage which, apart from an inlet for the starting reaction gas mixture, optionally further inlets for auxiliary gases, and an outlet for the product gas mixture, is sealed on the gas side, in the reaction stage directly oxidizing the propane and/or isobutane present in the starting reaction gas mixture partially to at least one target product by passing the starting reaction gas mixture at elevated temperature over a solid state catalyst, and conducting the reaction gas mixture as a product gas mixture comprising at least one target product and having the outlet pressure P^2 out of the reaction stage and, with this pressure P^2 , into a workup stage which, apart from an inlet for the product gas mixture, optionally further inlets for auxiliary gases, and an outlet for the residual product gas mixture, is sealed on the gas side, in the workup stage basically separating target product present in the product gas mixture of the reaction stage from said product gas mixture into a liquid phase and conducting the remaining residual product gas mixture which comprises not only propane and/or isobutane and also in some cases propene and/or isobutene and has the outlet pressure P^3 , where $P^3 < P^1$, out of the workup stage and recycling propane and/or isobutane present in

the residual product gas mixture into the reaction stage, which comprises selecting P^1 in such a way that $P^3 \geq 1.5$ bar and dividing the residual product gas mixture into two portions of the same composition and discharging one portion as output and recycling the other portion as cycle gas and feeding it back to the reaction stage, compressed to the inlet pressure P^1 , as a constituent of the starting reaction gas mixture.

See original Claim 1, and the specification at page 1, prenumbered lines 4-23.

VI. GROUNDS OF REJECTION

Ground (A)

Claims 1-32 stand rejected under 35 U.S.C. § 103(a) as unpatentable over U.S. 5,380,933 (Ushikubo et al) in view of EP 1193240 (Bogan et al).

Ground (B)

Claims 1-14 and 17-32 stand rejected under 35 U.S.C. § 112, first paragraph, as failing to satisfy the enablement requirement thereof

VII. ARGUMENT

Ground (A)

Claims 1-32 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Ushikubo et al in view of Bogan et al. That rejection is untenable and should not be sustained.

As recited in Claim 1 herein, the present invention is a process for heterogeneously catalyzed partial direct oxidation of propane and/or isobutane to at least one of the target products acrylic acid, methacrylic acid, by feeding a starting reaction gas mixture comprising propane and/or isobutane, molecular oxygen and at least one inert diluent gas and having a

inlet pressure P^1 to a reaction stage which, apart from an inlet for the starting reaction gas mixture, optionally further inlets for auxiliary gases, and an outlet for the product gas mixture, is sealed on the gas side, in the reaction stage directly oxidizing the propane and/or isobutane present in the starting reaction gas mixture partially to at least one target product by passing the starting reaction gas mixture at elevated temperature over a solid state catalyst, and conducting the reaction gas mixture as a product gas mixture comprising at least one target product and having the outlet pressure P^2 out of the reaction stage and, with this pressure P^2 , into a workup stage which, apart from an inlet for the product gas mixture, optionally further inlets for auxiliary gases, and an outlet for the residual product gas mixture, is sealed on the gas side, in the workup stage basically separating target product present in the product gas mixture of the reaction stage from said product gas mixture into a liquid phase and conducting the remaining residual product gas mixture which comprises not only propane and/or isobutane and also in some cases propene and/or isobutene and has the outlet pressure P^3 , where $P^3 < P^1$, out of the workup stage and recycling propane and/or isobutane present in the residual product gas mixture into the reaction stage, which comprises selecting P^1 in such a way that $P^3 \geq 1.5$ bar and dividing the residual product gas mixture into two portions of the same composition and discharging one portion as output and recycling the other portion as cycle gas and feeding it back to the reaction stage, compressed to the inlet pressure P^1 , as a constituent of the starting reaction gas mixture.

The key features of the presently-claimed process are as follows:

1. After separating the target product from the product gas mixture, no further separation of the propane and/or isobutane from the gas mixture is carried out, but the remaining mixture is simply divided into two portions of identical composition and one of these portions is recycled to the reaction zone.
2. The reaction is carried out at an inlet pressure, $P^1 > 1.5$ bar.

3. The outlet pressure of the workup stage, $P^3 \geq 1.5$ bar.
4. The outlet pressure of the reaction stage, P^2 , is the inlet pressure for the workup stage.

An important precondition for fulfillment of above features 2 and 3 is feature 1. The claimed process is beneficial due to recycling the propane and/or isobutane-containing residual product gas mixture that remains after separating the target product from the product gas mixture without previously separating propane and/or isobutane in the process. By avoiding such early separation, significant pressure losses, resulting in costly compression requirements, are avoided.

The applied prior art neither discloses nor suggests the above-discussed features. Rather, Ushikubo et al simply discloses the basic vapor phase catalytic oxidation of, for example, propane or isobutane to form the corresponding unsaturated carboxylic acid in the presence of a catalyst, wherein the invention in Ushikubo et al is drawn to the amount range of essential components of the catalyst. Bogan et al is similar to Ushikubo et al but uses a different (but somewhat similar) catalyst. The Examiner relies on Bogan et al for its disclosure of reaction zone pressure of from 0 to 75 psig [0065] as a basis for carrying out the reaction in Ushikubo et al at this pressure.

In reply, the Examiner has apparently overlooked the disclosure in Ushikubo et al that their reaction can be conducted usually under atmospheric pressure, but may be conducted under a slightly elevated pressure or slightly reduced pressure (sentence bridging columns 5 and 6). Nevertheless, the present invention is not simply carrying out a conventional heterogeneously catalyst partial direct oxidation of propane and/or isobutane at a conventional pressure. Rather, the present invention is directed to the above-discussed features. Thus, Ushikubo et al discloses and suggests nothing that would inform a person of ordinary skill in the art that high pressure is beneficial in the reaction stage, or that any

recycling of raw material would be beneficial, nor does it provide any hint of at which pressure the workup of the product gas should be carried out or what the outlet pressure of the worked-up product should be. Similarly, Bogan et al is silent with regard to pressure during purification or at its outlet.

In response to the above argument, the Examiner responds as follows:

Furthermore, the control of either the inlet or outlet pressure of the reaction zone is within the purview of the skilled artisan in the art; in addition, the limitation of the process with respect to the pressure ranges of the inlet or outlet pressure does not have [sic] impart any patentability to a process when such values are those which would be determined by one of the skilled artisan [sic] in the art in achieving optimum operation of the process. Also, any recycling of raw material is well-known in the art to make the process economical. Therefore, it would have been obvious to the skilled artisan in the art to control either the inlet or outlet pressure of the reaction zone so as to achieve the optimum operation of the process in the absence of an unexpected result.

In reply, and as discussed above, the present invention is not simply control of either the inlet or outlet pressure of the reaction zone, nor is the relationship between the three pressures recited, P^1 through P^3 , a known result-effective variable. It is not obvious to optimize a variable not known to be result-effective. *In re Antonie*, 559 F.2d 618, 195 USPQ 6, 8-9 (CCPA 1977) (exceptions to rule that optimization of a result-effective variable is obvious, such as where the variable was not recognized to be result effective). Moreover, while recycling of raw material may be well-known in general, the present invention does not involve simply recycling *per se*.

For all the above reasons, it is respectfully requested that this rejection be REVERSED.

Ground (B)

Claims 1-14 and 17-32 stand rejected under 35 U.S.C. § 112, first paragraph, as failing to satisfy the enablement requirement thereof. That rejection is untenable and should not be sustained.

It should be clear from the discussion of the prior art rejection, above, that the present invention is based on various pressures at various points in the process, combined with separation and recycle of product mixtures. The invention is not limited to particular catalysts and indeed, the number of catalysts known for heterogeneously catalyzed partial direct oxidation of propane and/or isobutane to form (meth)acrylic acid is vast. It is clear from the specification as a whole that the presently-claimed invention could be carried out with any such catalyst. Indeed, Applicants disclose in the specification at page 9, lines 27-29 that “[u]seful catalysts for the process according to the invention are in principle all of those which are recommended in the prior art for the heterogeneously catalyzed partial direct oxidation of propane and/or isobutane to at least one of the target products.”

In response to the above argument, the Examiner cites the so-called *Wands* factors, but as pointed out above, the present invention is not limited to particular catalysts, and one skilled in the art would be enabled to choose applicable catalysts by routine experimentation.

For all the above reasons, it is respectfully requested that this rejection be REVERSED.

VIII. CONCLUSION

For the above reasons, it is respectfully requested that all the rejections still pending in the Final Office Action be REVERSED.

Respectfully submitted,

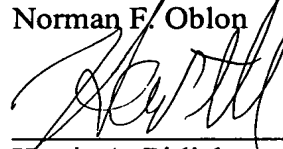
Customer Number

22850

Tel: (703) 413-3000
Fax: (703) 413 -2220
(OSMMN 06/04)

OBLON, SPIVAK, McCLELLAND,
MAIER & NEUSTADT, P.C.

Norman F. Oblon



Harris A. Pitlick

Registration No. 38,779

NFO:HAP\



CLAIMS APPENDIX

1. A process for heterogeneously catalyzed partial direct oxidation of propane and/or isobutane to at least one of the target products acrylic acid, methacrylic acid, by feeding a starting reaction gas mixture comprising propane and/or isobutane, molecular oxygen and at least one inert diluent gas and having an inlet pressure P^1 to a reaction stage which, apart from an inlet for the starting reaction gas mixture, optionally further inlets for auxiliary gases, and an outlet for the product gas mixture, is sealed on the gas side, in the reaction stage directly oxidizing the propane and/or isobutane present in the starting reaction gas mixture partially to at least one target product by passing the starting reaction gas mixture at elevated temperature over a solid state catalyst, and conducting the reaction gas mixture as a product gas mixture comprising at least one target product and having the outlet pressure P^2 out of the reaction stage and, with this pressure P^2 , into a workup stage which, apart from an inlet for the product gas mixture, optionally further inlets for auxiliary gases, and an outlet for the residual product gas mixture, is sealed on the gas side, in the workup stage basically separating target product present in the product gas mixture of the reaction stage from said product gas mixture into a liquid phase and conducting the remaining residual product gas mixture which comprises not only propane and/or isobutane and also in some cases propene and/or isobutene and has the outlet pressure P^3 , where $P^3 < P^1$, out of the workup stage and recycling propane and/or isobutane present in the residual product gas mixture into the reaction stage, which comprises selecting P^1 in such a way that $P^3 \geq 1.5$ bar and dividing the residual product gas mixture into two portions of the same composition and discharging one portion as output and recycling the other portion as cycle gas and feeding it back to the reaction stage, compressed to the inlet pressure P^1 , as a constituent of the starting reaction gas mixture.

2. The process as claimed in claim 1, wherein the residual product gas mixture contains at least 5% by volume of constituents other than propane and/or isobutane and also other than propene and/or isobutene.

3. The process as claimed in claim 1, wherein the residual product gas mixture contains at least 10% by volume of constituents other than propane and/or isobutane and also other than propene and/or isobutene.

4. The process as claimed in claim 1, wherein the pressure $P^3 \geq 1.5$ bar and ≤ 25 bar.

5. The process as claimed in claim 1, wherein the pressure $P^3 \geq 1.5$ bar and ≤ 20 bar.

6. The process as claimed in claim 1, wherein the pressure $P^3 \geq 1.5$ bar and ≤ 10 bar.

7. The process as claimed in claim 1, wherein the pressure $P^3 \geq 2$ bar and ≤ 8 bar.

8. The process as claimed in claim 1, wherein the pressure P^1 is from 1 to 4 bar above the pressure P^3 .

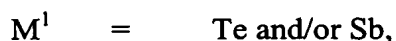
9. The process as claimed in claim 1, wherein the pressure P^1 is from 1.5 to 3.5 bar above the pressure P^3 .

10. The process as claimed in claim 1, wherein P^1 is from 3 to 10 bar.

11. The process as claimed in claim 1, wherein P^1 is from 4 to 8 bar.
12. The process as claimed in claim 1, wherein the portion of the residual product gas mixture which is discharged as output is discharged via an expander.
13. The process as claimed in claim 1, wherein the reaction stage is a catalyst-charged tube bundle reactor or fluidized bed reactor.
14. The process as claimed in claim 1, wherein the workup stage is an absorption column or a column for fractional condensation or a series arrangement of quench stages.
15. The process as claimed in claim 1, wherein the active composition of the catalyst is a multimetal oxide composition which comprises the elements Mo, V, at least one of the two elements Te and Sb, and at least one of the elements from the group consisting of Nb, Ta, W, Ti, Al, Zr, Cr, Mn, Ga, Fe, Ru, Co, Cs, Ca, Sr, Ba, Rh, Ni, Pd, Pt, La, Pb, Cu, Re, Ir, Y, Pr, Nd, Tb, Bi, B, Ce, Sn, Zn, Si, Na, Li, K, Mg, Ag, Au and In in combination.
16. The process as claimed in claim 1, wherein the active composition of the catalyst is a multimetal oxide composition which contains the element combination having the stoichiometry I



where



M^2 = at least one of the elements from the group consisting of Nb, Ta, W, Ti, Al, Zr, Cs, Ca, Sr, Ba, Cr, Mn, Ga, Fe, Ru, Co, Rh, Ni, Pd, Pt, La, Bi, Pb, Cu, Re, Ir, Y, Pr, Nd, Tb, Ce, Sn, Zn, Si, Na, Li, K, Mg, Ag, Au and In,

b = from 0.01 to 1

c = from > 0 to 1 and

d = from > 0 to 1.

17. The process as claimed in claim 1, wherein the oxygen source used is air.

18. The process as claimed in claim 1, wherein the reaction temperature is from 200 to 700°C.

19. The process as claimed in claim 1, wherein the starting reaction gas mixture contains

from 0.5 to 15% by volume of propane or isobutane,
from 10 to 90% by volume of air,
from 0 to 50% by volume of steam and
a remainder of cycle gas.

20. The process as claimed in claim 1, wherein the starting reaction gas mixture contains

from 0.5 to 15% by volume of propane or isobutane,
from 10 to 90% by volume of air,
from 10 to 50% by volume of steam and
a remainder of cycle gas.

21. The process as claimed in claim 1, wherein the starting reaction gas mixture contains

from 70 to 90% by volume of propane or isobutane,

from 5 to 25% by volume of molecular oxygen,

from 0 to 25% by volume of steam and

a remainder of cycle gas.

22. The process as claimed in claim 1, wherein the conversion from propane and/or isobutane, based on single pass of the reaction gas mixture through the reaction stage, is from 10 to 70 mol%.

23. The process as claimed in claim 22, wherein the selectivity of the target product formation is from 40 to 98 mol%.

24. The process as claimed in claim 1, wherein the target product present in the product gas mixture of the reaction stage is basically separated into the liquid phase in such a way that the molar ratio W of the steam present in the remaining residual product gas mixture to the propane present therein is at least 50% smaller than the corresponding molar ratio W' in the product gas mixture of the reaction stage.

25. The process as claimed in claim 1, wherein the target product present in the product gas mixture of the reaction stage is basically separated into the liquid phase in an absorption column by absorption into an organic solvent in such a way that the discharge from the absorption column is monophasic.

26. The process as claimed in claim 1, wherein the propane and/or isobutane and also any propene and/or isobutene present in the portion of the residual product gas mixture which is discharged as output are removed from said residual product gas mixture and recycled into the reaction stage, recompressed to the inlet pressure P^1 .

27. The process as claimed in claim 1, wherein the ratio V of that portion of the residual product gas mixture which is recycled as cycle gas to that portion of the residual product gas mixture which is discharged as output is ≥ 0.5 and ≤ 30 .

28. The process as claimed in claim 1, wherein the ratio V of that portion of the residual product gas mixture which is recycled as cycle gas to that portion of the residual product gas mixture which is discharged as output is ≥ 2 and ≤ 25 .

29. The process as claimed in claim 1, wherein the ratio V of that portion of the residual product gas mixture which is recycled as cycle gas to that portion of the residual product gas mixture which is discharged as output is ≥ 3 and ≤ 20 .

30. The process as claimed in claim 1, wherein the cycle gas is recompressed to the inlet pressure P^1 using a blower.

31. The process as claimed in claim 1, wherein the oxygen source used is air which is compressed to the inlet pressure P^1 by means of a radial compressor.

32. The process as claimed in claim 1, which is a process for partial direct oxidation of propane to acrylic acid.

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EVIDENCE APPENDIX

None.

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RELATED PROCEEDINGS APPENDIX

None.